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(54) **Polyamide resin composition.**

(57) There is disclosed a polyamide resin composition comprising (A) 100 parts by weight of a polyamide (a) composed mainly of a xylenediamine component and an α,ω -straight chain aliphatic dibasic acid component, or a combination of the polyamide (a) and polyamide 66, (B) 1 to 15 parts by weight of polyamide 12, (C) 0.01 to 5 parts by weight (in terms of copper) of a copper compound, (D) 1 to 15 parts by weight of carbon black, and (E) an alkali metal halide of such an amount that the number of halogen atoms of the alkali metal halide is 0.3 to 4 per one copper atom of the above copper compound. Said polyamide resin composition has an excellent weather resistance, moldability, and mechanical properties, and can be injection-molded.

EP 0 580 387 A1

Background of the InventionField of the Invention

5 The present invention relates to a polyamide resin composition. More particularly, the present invention relates to a polyamide resin composition which is excellent in weather resistance, moldability and mechanical properties, and which can be injection-molded.

Prior Art

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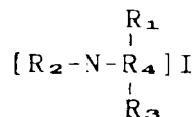
Polyamide resins have good mechanical properties, moldability and chemical resistance and are in wide use as automobile parts, machine parts, building materials, etc. Polyamide resins, however, are very susceptible to oxidative deterioration and, when deteriorated by oxidation, reduce the polymerization degree. This reduction in polymerization degree invites undesirable phenomena such as reduction in mechanical properties, occurrence of surface cracks, coloring and the like. The oxidative deterioration is accelerated by heat or light and, therefore, use of polyamide resin in outdoor exposure is restricted.

In order to prevent the above oxidative deterioration, i.e. to improve the weather resistance of polyamide resin, addition of various stabilizers to polyamide resin is disclosed in Japanese Patent Application Kokai (Laid-Open) No. 93652/1973 and Japanese Patent Publication No. 29823/1989.

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That is, Japanese Patent Application Kokai (Laid-Open) No. 93652/1973 discloses a polyamide resin composition having excellent heat resistance and light resistance, obtained by adding to a polyamide a metal complex of a tetraalkylammonium iodide represented by the following general formula

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wherein R_1 , R_2 , R_3 and R_4 are each a hydrogen atom or an alkyl group having 1 to 5 carbon atoms and at least one of R_1 to R_4 is an alkyl group.

Japanese Patent Publication No. 29823/1989 discloses a reinforced polyamide molding material comprising (A) 0.01 to 5% by weight of carbon black, (B) 1 to 70% by weight of a glass fiber, and (C) 25 to 98.09% by weight of a polyamide per 100 % by weight of the total amount of the components (A), (B) and (C), wherein the particle size of the carbon black is such that the maximum particle diameter is 20 μm or less and the volume average particle diameter of those particles having diameters of 1 μm or more is 5 μm or less.

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The improvement of weather resistance by the above composition or material, however, is not satisfactory, and further improvement is desired.

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Summary of the Invention

An object of the present invention is to provide a polyamide resin composition which can be used in injection molding.

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Another object of the present invention is to provide a polyamide resin composition having excellent weather resistance.

A further object of the present invention is to provide a polyamide resin composition having excellent weather resistance and a balance in moldability and mechanical properties.

According to the present invention there is provided a polyamide resin composition comprising

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(A) 100 parts by weight of a polyamide (a) composed mainly of a xylylenediamine component and an α,ω -straight chain aliphatic dibasic acid component, or a combination of the polyamide (a) and polyamide 66, (B) 1 to 15 parts by weight of polyamide 12,

(C) 0.01 to 5 parts by weight (in terms of copper) of a copper compound,

(D) 1 to 15 parts by weight of carbon black, and

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(E) an alkali metal halide of such an amount that the number of halogen atoms of the alkali metal halide is 0.3 to 4 per one copper atom of the above copper compound.

The above objects can be achieved by this polyamide resin composition.

Detailed Description of the Invention

The polyamide resin composition of the present invention comprises a polyamide (a) composed mainly of a xylylenediamine component and an α,ω -straight chain aliphatic dibasic acid component, or a combination of the polyamide (a) and polyamide 66 [said polyamide (a) or said combination is hereinafter referred to as "resin component" sometimes].

The xylylenediamine component constituting the polyamide (a) includes m-xylylenediamine and p-xylylenediamine. The polyamide (a) may contain, besides the xylylenediamine component, an aliphatic diamine component or an aromatic diamine component in an amount of 30% by weight or less, preferably 20% by weight or less, based on the total diamine components. Preferably, m-xylylenediamine component is contained in an amount of 60% by weight or more, particularly 70% by weight or more, based on the total diamine components.

The α,ω -straight chain aliphatic dibasic acid component preferably has 6 to 12 carbon atoms, and examples thereof include adipic acid, sebacic acid, suberic acid, dodecanedioic acid and eicosanoic acid. Of these, adipic acid is preferable in view of the moldability and mechanical properties required for the polyamide resin composition of the present invention.

The polyamide (a) may contain a dibasic acid component other than the α,ω -straight chain aliphatic dibasic acid component, such as aromatic dibasic acid, chain aliphatic dibasic acid or the like in an amount of 30% by weight or less, preferably 20% by weight or less, based on the total amount of the dibasic components.

The polyamide (a) used in the present invention preferably has a relative viscosity of 1.8 to 3.0, particularly 2.0 to 2.4, as measured at 25°C for a solution of 1 g of the polyamide (a) dissolved in 100 ml of 98% sulfuric acid.

Such a polyamide (a) is a substance per se known, and can be easily produced by conducting polycondensation by a known process using m-xylylenediamine and an α,ω -straight chain aliphatic dibasic acid as main starting materials.

Polyamide 66 is a known polyamide composed mainly of hexamethylenediamine and adipic acid.

When the polyamide (a) and polyamide 66 are used in combination, the polyamide resin composition of the present invention can have improved moldability and the cycle time of injection molding can be shortened, as compared with when the polyamide (a) alone is used.

When the polyamide (a) and polyamide 66 are used in combination, the proportion of polyamide 66 is preferably 40% by weight or less, more preferably 1 to 40% by weight, particularly preferably 3 to 40% by weight, in view of the mechanical strengths and low water absorption required for the present polyamide resin composition.

The polyamide resin composition of the present invention comprises polyamide 12, a copper compound, carbon black and an alkali metal halide for the purpose of improving weather resistance.

Polyamide 12 is a substance per se known, and preferably has a melt flow rate of 20 to 250 g/10 min, particularly 30 to 100 g/10 min., as measured in accordance with ASTM D 1238 under the conditions of the load of 10 kg at a temperature of 275°C. When the melt flow rate of polyamide 12 is in the above range, the resulting polyamide resin composition exhibits good weather resistance and excellent mechanical strengths.

Polyamide 12 is used preferably in an amount of 1 to 15 parts by weight, particularly 5 to 10 parts by weight per 100 parts by weight of the resin components. When the amount of polyamide 12 is in the above range, the polyamide resin composition of the present invention has improved weather resistance and appropriate levels of mechanical properties.

The copper compound is selected from copper halides such as copper chloride, copper bromide, copper fluoride and copper iodide; copper salts of carboxylic acids having 2 to 18 carbon atoms, such as copper acetate, copper naphthenate, copper caprate, copper laurate and copper stearate; copper thiocyanate; copper nitrate; copper acetylacetonate; copper (I) oxide; and copper (II) oxide. Among these compounds, copper halides, copper (I) oxide and copper (II) oxide are preferably used. The above compounds can be used singly or in combination.

The amount of the copper compound used is preferably at least 0.01 part by weight, more preferably 0.01 to 5 parts by weight, particularly preferably 0.01 to 1.5 parts by weight, in terms of copper, per 100 parts by weight of the resin components, for improving weather resistance of the polyamide resin composition.

The alkali metal halide includes a fluoride, bromide or iodide of lithium, sodium or potassium. Of them, potassium iodide is preferable. They can be used singly or in combination. The use of such an alkali metal halide improves the dispersion of the copper compound in the polyamide resin composition and consequently, the weather resistance of the composition is improved.

The alkali metal halide is used preferably in such an amount that the number of halogen atoms of the alkali metal halide is 0.3 to 4, particularly 0.4 to 2.5 per one copper atom of the copper compound.

The carbon black used in the present invention preferably has an average particle diameter of 17 to 27

μm, particularly 19 to 25 μm for the maintenance of good dispersion in the composition, and further preferably, it has an oil absorption of 50 to 100 cc/100 g, particularly 53 to 75 cc/100 g. The above oil absorption is a value obtained by measurement described in JIS K 6221 using dibutyl phthalate.

Carbon black is used in an amount of 1 to 10 parts by weight, preferably 3 to 5 parts by weight per 100 parts by weight of the resin components for improvement of weather resistance of the polyamide resin composition and the maintenance of appropriate levels of mechanical properties of the composition.

The polyamide resin composition of the present invention may comprise at least one plastics or elastomer selected from polyethylene terephthalate, polybutylene terephthalate, polycarbonate, polyphenylene ether, polyphenylene sulfide, AS resin, ABS resin, polyolefins, ethylene-propylene rubber, etc. in such an amount that the objects of the present invention are not impaired. It is desirable that the amount of the plastics or elastomer added is 15 parts by weight, particularly 10 parts by weight, at the most per 100 parts by weight of the resin component.

The polyamide resin composition of the present invention may further comprise, as required, a reinforcing fiber material such as glass fiber, carbon fiber or the like for improvement of the mechanical strengths.

The amount of the reinforcing fiber material used is preferably 30 to 140 parts by weight per 100 parts by weight of the resin component.

The polyamide resin composition of the present invention may further comprise, as required, a filler such as mica or the like for improvement of the stiffness.

The amount of the filler used is preferably 20 to 80 parts by weight per 100 parts by weight of the resin component.

The reinforcing fiber material and the filler may be used singly or in combination.

The polyamide resin composition of the present invention may further comprise, as required, additives such as a nucleating agent, a plasticizer, a mold-release agent, an antistatic agent, a flame retardant, a lubricant and a pigment.

The polyamide resin composition of the present invention can be generally produced by melt-kneading components using an extruder, at a temperature higher, by 5 to 50°C, than the melting point or the flow-starting temperature of polyamide (a), followed by pelletization.

The polyamide resin composition of the present invention can be molded, by, for example, injection molding, into automobile exterior trim parts and building materials usable in an uncoated state, for example, automobile door mirror stay, fashion rail, door handle and handrail parts.

The present invention is hereinafter described in more detail by way of Examples.

The test pieces prepared in Examples and Comparative Examples were measured for the following test items by the following test methods.

(1) Mechanical properties

Tensile strength: measured by ASTM D 638.

Tensile modulus: measured by ASTM D 638.

Flexural strength: measured by ASTM D 790.

Flexural modulus: measured by ASTM D 790.

(2) Color tone

The L value and color tone change of a test piece subjected to an exposure test were measured using a color difference meter (Model: Z-Σ80 manufactured by Nihon Denshoku Kogyo K.K.).

(3) Occurrence of surface cracks

The occurrence of surface cracks of a test piece that was subjected to an exposure test was observed using a polarization microscope (model: OPTIPHOT-POL manufactured by NIPPON KOGAKU K.K.).

The following rating standards were employed in Tables 1 and 2.

○: No surface cracks were observed.

X: Surface cracks were observed.

(4) Surface gloss

The change of surface gloss of a test piece subjected to an exposure test was rated visually. The following rating standards were employed in Tables 1 and 2.

○: The color tone and gloss after exposure were nearly the same as before exposure.

Δ: The color tone and gloss after exposure were slightly inferior to those before exposure.

X: The color tone and gloss after exposure were inferior to those before exposure.

Example 1

The following components were mixed with a tumbler to obtain a mixture.

● 90 parts by weight of a poly(m-xylyleneadipamide) (hereinafter referred to as "MX nylon") manufactured

by MITSUBISHI GAS CHEMICAL COMPANY, INC. having a relative viscosity as measured at 25°C for a solution of 1 g of said polymer dissolved in 100 ml of 98% sulfuric acid (said relative viscosity is hereinafter referred to as "relative viscosity"), of 2.14.

- 5 parts by weight of polyamide 66 of pellet shape having a relative viscosity of 2.25.
- 5 parts by weight of polyamide 12 of pellet shape having a melt flow rate of 80 g/10 min.
- 0.2 part by weight (0.067 part by weight in terms of copper) of copper iodide.
- 0.2 part by weight of potassium iodide (I/Cu molar ratio = 2.15).
- 5 parts by weight of carbon black (#45, manufactured by Mitsubishi Chemical Industries, Ltd.) having an average particle diameter of 24 μm and an oil absorption of 53 cc/100 g.

The mixture was melt-kneaded at 275°C using a vented extruder, and then extruded into strands. The strands were cooled in a water bath, cut and dried to prepare a pellet-shaped molding material. This molding material was injection-molded at a mold temperature of 130°C to obtain test pieces.

Some of the test pieces were measured for the above-mentioned mechanical properties (1) and color tone (2).

Other test pieces were subjected to an exposure test.

The exposure test was conducted by placing a test piece in a sunshine weather meter (model: WEL-SUN-HC manufactured by Suga Shikenki K.K.) and subjecting the test piece to light exposure and also to water spraying (12 minutes per hour) while maintaining the black panel temperature at 63°C.

The test pieces after 500-hour or 1,000-hour exposure were measured for the above-mentioned test items (1) to (4). The results are shown in Table 1.

Example 2

The procedure of Example 1 was repeated with the exception that the amount of carbon black used was changed to 3 parts by weight. The results are shown in Table 1.

Example 3

40 Parts by weight of the same MX nylon as used in Example 1, 5 parts by weight of the same polyamide 66 as used in Example 1, 5 parts by weight of the same polyamide 12 as used in Example 1, 50 parts by weight of glass fiber-chopped strands of 3 mm in length [CS03JAFT2 (trade name) manufactured by Asahi Fiber Glass Co., Ltd], 0.2 part by weight of copper iodide, 0.2 part by weight of potassium iodide, and 5 parts by weight of the same carbon black as used in Example 1 were melt-kneaded, extruded into strands, cooled, cut and dried in the same manner as in Example 1 to obtain a pellet-shaped molding material. The material was measured for weather resistance in the same manner as in Example 1. The results are shown in Table 1.

Example 4

The procedure of Example 3 was repeated with the exception that 0.2 part by weight of copper (I) oxide was used in place of copper iodide. The results are shown in Table 1.

Example 5

The procedure of Example 3 was repeated with the exception that the amount of the glass fiber-chopped strands used was changed from 50 parts by weight to 30 parts by weight and further 20 parts by weight of a mica powder having an average particle diameter of 90 μm [B-82 (trade name), manufactured by Yamaguchi Mica K.K.] was used. The results are shown in Table 1.

Examples 6 and 7

The procedure of Example 3 was repeated with the exception that there was used, as polyamide 12, one having a melt flow rate of 250 g/10 min (Example 6) or 35 g/10 min (Example 7). The results are shown in Table 1.

Comparative Example 1

The procedure of Example 1 was repeated with the exception that no polyamide 12 was used. The results are shown in Table 2.

Comparative Example 2

The procedure of Example 1 was repeated with the exception that neither copper iodide nor potassium iodide was used. The results are shown in Table 2.

Comparative Example 3

The procedure of Example 1 was repeated with the exception that no carbon black was used. The results are shown in Table 2.

Comparative Example 4

The procedure of Example 4 was repeated with the exception that no polyamide 12 was used. The results are shown in Table 2.

As is clear from the above Examples and Comparative Examples, the polyamide resin composition of the present invention has excellent weather resistance.

Table 1

Example No.	1	2	3	4
Formulation (wt.parts)				
MX nylon	90	90	40	40
Polyamide 66	5	5	5	5
Polyamide 12	5	5	5	5
Glass fiber	-	-	50	50
Mica	-	-	-	-
Copper iodide	0.2	0.2	0.2	-
Copper (I) oxide	-	-	-	0.2
Potassium iodide	0.2	0.2	0.2	0.2
Carbon black	5	3	5	5
Cu*	0.07	0.07	0.15	0.39
I/Cu**	2.15	2.15	2.15	0.43
Before exposure				
Mechanical properties				
Tensile strength (kgf/cm ²)	800	850	2320	2350
Tensile elongation (%)	2.0	2.0	1.6	1.6
Flexural strength (kgf/cm ²)	1600	1650	3170	3200
Flexural modulus (10 ³ kgf/cm ²)	45	45	160	160
Color tone				
L value	9.79	10.02	10.42	10.48
Color difference ΔE	0	0	0	0
After 500-hour exposure				
Mechanical properties				
Tensile strength (kgf/cm ²)	780	820	2270	2290
Tensile elongation (%)	1.9	1.9	1.5	1.5
Flexural strength (kgf/cm ²)	1560	1600	3100	3120
Flexural modulus (10 ³ kgf/cm ²)	44	44	160	160
Color tone				
L value	12.20	12.72	13.99	14.01
Color difference ΔE	3.14	3.30	3.86	3.88
Occurrence of surface cracks	○	○	○	○
Surface gloss (visual check)	○	○	○	○
After 1,000-hour exposure				
Mechanical properties				
Tensile strength (kgf/cm ²)	760	800	2220	2230
Tensile elongation (%)	1.8	1.8	1.4	1.4
Flexural strength (kgf/cm ²)	1520	1570	3080	3100
Flexural modulus (10 ³ kgf/cm ²)	43	43	158	158
Color tone				
L value	14.36	14.58	15.58	15.82
Color difference ΔE	4.52	4.72	5.33	5.70
Occurrence of surface cracks	○	○	○	○
Surface gloss (visual check)	○	○	○	○

- to be continued -

Table 1 (continued)

Example No.	5	6	7
Formulation (wt. parts)			
MX nylon	40	40	40
Polyamide 66	5	5	5
Polyamide 12	5	5	5
Glass fiber	30	50	50
Mica	20	-	-
Copper iodide	0.2	0.2	0.2
Copper (I) oxide	-	-	-
Potassium iodide	0.2	0.2	0.2
Carbon black	5	5	5
Cu*	0.15	0.15	0.15
I/Cu**	2.15	2.15	2.15
Before exposure			
Mechanical properties			
Tensile strength (kgf/cm ²)	1800	2320	2320
Tensile elongation (%)	1.0	1.6	1.6
Flexural strength (kgf/cm ²)	2400	3170	3170
Flexural modulus (10 ³ kgf/cm ²)	200	160	160
Color tone			
L value	10.12	10.42	10.42
Color difference ΔE	0	0	0
After 500-hour exposure			
Mechanical properties			
Tensile strength (kgf/cm ²)	1780	2270	2270
Tensile elongation (%)	1.0	1.5	1.5
Flexural strength (kgf/cm ²)	2350	3100	3100
Flexural modulus (10 ³ kgf/cm ²)	197	160	160
Color tone			
L value	14.10	13.80	14.90
Color difference ΔE	4.00	3.50	4.80
Occurrence of surface cracks	○	○	○
Surface gloss (visual check)	○	○	○
After 1,000-hour exposure			
Mechanical properties			
Tensile strength (kgf/cm ²)	1750	2220	2220
Tensile elongation (%)	0.9	1.4	1.4
Flexural strength (kgf/cm ²)	2300	3080	3080
Flexural modulus (10 ³ kgf/cm ²)	195	158	158
Color tone			
L value	16.00	15.20	17.00
Color difference ΔE	5.90	5.00	6.80
Occurrence of surface cracks	○	○	○
Surface gloss (visual check)	○	○	○

Table 2

Comparative Example No.	1	2	3	4
Formulation (wt.parts)				
MX nylon	95	90	90	45
Polyamide 66	5	5	5	5
Polyamide 12	-	5	5	-
Glass fiber	-	-	-	50
Copper iodide	0.2	-	0.2	-
Copper (I) oxide	-	-	-	0.2
Potassium iodide	0.2	-	0.2	0.2
Carbon black	5	5	-	5
Cu*	0.067	-	0.067	0.36
I/Cu**	2.15	-	2.15	0.43
Before exposure				
Mechanical properties				
Tensile strength (kgf/cm ²)	850	800	880	2350
Tensile elongation (%)	2.0	2.0	2.1	1.6
Flexural strength (kgf/cm ²)	1650	1600	1700	3150
Flexural modulus (10 ³ kgf/cm ²)	46	45	46	160
Color tone				
L value	9.85	9.82	-	10.50
Color difference ΔE	0	0	-	0
After 500-hour exposure				
Mechanical properties				
Tensile strength (kgf/cm ²)	790	730	780	2250
Tensile elongation (%)	1.9	1.9	2.0	1.5
Flexural strength (kgf/cm ²)	1570	1500	1600	3100
Flexural modulus (10 ³ kgf/cm ²)	44	44	45	160
Color tone				
L value	18.52	19.63	-	17.49
Color difference ΔE	7.68	9.22	-	7.59
Occurrence of surface cracks	○	X	X	○
Surface gloss (visual check)	△	△	△	△
After 1,000-hour exposure				
Mechanical properties				
Tensile strength (kgf/cm ²)	700	680	740	2170
Tensile elongation (%)	1.7	1.7	1.7	1.4
Flexural strength (kgf/cm ²)	1500	1450	1500	1960
Flexural modulus (10 ³ kgf/cm ²)	42	42	42	157
Color tone				
L value	21.56	21.07	-	20.07
Color difference ΔE	10.22	10.03	-	9.71
Occurrence of surface cracks	X	X	X	X
Surface gloss (visual check)	X	X	X	X

(Note) In the above Tables 1 and 2, the symbols "*" and "**" are defined as follows:

*: Proportion of copper of copper iodide or copper (I) oxide per 100 parts by weight of the total of MX nylon and polyamide 66.

**: The number of iode atoms per one copper atom of the copper compound.

Claims

1. A polyamide resin composition comprising
 - (A) 100 parts by weight of a polyamide (a) composed mainly of a xylylenediamine component and an α,ω -straight chain aliphatic dibasic acid component, or a combination of the polyamide (a) and polyamide 66,
 - (B) 1 to 15 parts by weight of polyamide 12,
 - (C) 0.01 to 5 parts by weight (in terms of copper) of a copper compound,
 - (D) 1 to 15 parts by weight of carbon black, and
 - (E) an alkali metal halide of such an amount that the number of halogen atoms of the alkali metal halide is 0.3 to 4 per one copper atom of the above copper compound.
2. A polyamide resin composition according to Claim 1, wherein the combination of the polyamide (a) and polyamide 66 contains polyamide 66 in an amount of 40% by weight or less.
3. A polyamide resin composition according to Claim 1, which further comprises 30 to 140 parts by weight of a reinforcing fiber material per 100 parts by weight of the polyamide (a) or the combination of the polyamide (a) and polyamide 66.
4. A polyamide resin composition according to Claim 1, which further comprises 20 to 80 parts by weight of mica per 100 parts by weight of the polyamide (a) or the combination of the polyamide (a) and polyamide 66.
5. A polyamide resin composition according to Claim 1, wherein the polyamide 12 has a melt flow rate of 20 to 250 g/10 min. as measured by ASTM D 1238 under the conditions of the load of 10 kg at a temperature of 275°C.
6. A polyamide resin composition according to Claim 1, wherein the copper compound is a copper halide, a copper (I) oxide or a copper (II) oxide.
7. A polyamide resin composition according to Claim 1, wherein the alkali metal halide is potassium iodide.
8. A polyamide resin composition according to Claim 1, wherein the α,ω -straight chain aliphatic dibasic acid component has 6 to 12 carbon atoms.
9. A polyamide resin composition according to Claim 1, which is suitable for injection molding.
10. An article obtained by injection-molding the polyamide resin composition of Claim 1.
11. Use of the polyamide resin composition of Claim 1 as a resin for injection molding.



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 93 30 5653

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	EP-A-2 596 567 (MITSUBISHI GAS) ---		C08L77/06 C08K3/00
A	EP-A-0 157 756 (MONSANTO) ---		/(C08L77/06 ,L77:02)
A	US-A-2 868 757 (N.K.J. SYMONS) ---		(C08K3/00 ,L77:00)
A	US-A-2 845 962 (D.B. ERDINGTON) ---		
A	CH-A-484 963 (BAYER) ---		
A	GB-A-1 200 454 (I.C.I.) ---		
A	US-A-3 558 553 (R.A. HAYES ET AL.) ---		
A	FR-A-2 300 782 (RHONE-POULENC) -----		
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C08L C08K
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 19 OCTOBER 1993	Examiner LEROY ALAIN
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			

EPO FORM 1303 03.92 (P.401)

